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### **III. REMARKS**

#### **RESPONSE TO REJECTIONS**

##### **In the Claims:**

Claim rejections under 35 U.S.C. 112:

##### ***Examiner's Position:***

Claims 31 and 32 rejected under 35 U.S.C. 112, first paragraph, as failing to comply with the written description requirement. The specification allegedly does not support the limitation of claim 31 and 32 (see the entire specification for details), namely "a mixture of alkyl aluminum halide and a zirconium(IV) alkoxide alcohol preparation". This compound is totally not supported from the specification.

Claims 31-32 are rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. It is unclear what "aliphaticor" on line 4 of claim 31 is. Regarding claim 31 and 32, the term "a zirconium(IV) alkoxide alcohol preparation" cannot be understood. Is it a compound, a process, or something else?

##### **Applicant's Response:**

Applicant respectfully traverses the alleged lack of disclosure of the instant Specification. On the contrary, the instant Specification is replete with a description of a

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“multi-component system catalyst essentially comprising zirconium alkoxide, alkyl aluminumhalide/alkylaluminums” (page 4, first paragraph). It is clear that the individual components of the inventive reaction are separately added but jointly in a reaction mixture during the catalytically facilitated oligomerization of ethylene to low molecular weight (LMW) olefins. However, Applicant has cancelled claim 31 to expedite the prosecution the presently claimed process; the rejection is therefore deemed moot. Moreover, the cancellation of rejected claim 31 renders any required correction moot concerning the inadvertent typographical error in the term “aliphaticor” which should have been spelled separately --aliphatic or--.

As described on page 4, and e.g. instant Example 4, line 3, e.g.,  $\text{Zr}(\text{OBu})_4 \cdot \text{BuOH}$  designates a catalyst component consisting of equal moles of  $\text{Zr}(\text{OBu})_4$  and BuOH. The amendment of claim 32 is therefore believed to overcome the rejection under the second paragraph of the statute, such that the component of step (ii) can consist of  $\text{Zr}(\text{OBu})_4 \cdot \text{BuOH}$ , clearly describing a zirconium compound prepared and added to the reactor in the presence of BuOH within the prescribed mole ratio parameters of 1:0.33 to 1:1.23.

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- **Claim Rejections under 35 U.S.C. § 103(a):**

- ***The Examiner's Position:***

Claims 1-3, 14-17, 18, 31, and 32 are rejected under 35 U.S.C. 103(a) as being obvious over Langer, Jr. (4,409,414) ("Langer"). In particular, the Examiner alleges that Langer discloses a batch/continuous process of making alpha-linear olefins having applicants' claimed range of number of carbons by oligomerizing ethylene in the presence of a catalyst containing zirconium alkoxide such as  $Zr(OBu)_4$  and alkyl aluminum halide, in the presence of a diluent such as toluene under the condition of a temperature ranging from below 125°C, a pressure ranging from above 50 psia, such as 500 psia, during the claimed time and high-speed stirring (the abstract; col. 2, line 18 thru col. 6, line 34; col. 7, lines 12-29; examples, and the entire reference for details). Moreover, the Examiner contends that Langer discloses the process as operated in the presence of alcohol to enhance the polymerization process (col. 5, lines 55-68), ratio of the amount alcohol and the alkyl group of aluminum alkyl (column 6, lines 1-4) and the amount of alcohol effects to the molecular weight of the product (col. 5, lines 55-57). It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by selecting an appropriate amount of the added alcohol, such as the claimed one according to the desired molecular weight of the product since it has been held by the patent law that the selection of reaction parameters such as temperature and concentration would have been obvious.

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More particularly, where the general conditions of the claimed are disclosed in the prior art, it is not inventive to discover the optimum or workable ranges by routine experimentation. In re Aller 105 USPQ 233, 255 (CCPA 1955). In re Waite 77 USPQ 586 (CCPA 1948). In re Scheri 70 USPQ 204 (CCPA 1946). In re Irmischer 66 USPQ 314 (CCPA 1945). In re Norman 66 USPQ 308 (CCPA 1945). In re Swenson 56 USPQ 372 (CCPA 1942). In re Sola 25 USPQ 433 (CCPA 1935). In re Dreyfus 24 USPQ 52 (CCPA 1934).

New claims recite steps of making the catalyst used for the process by adding components. However, it has been held that a proper sequence of adding ingredients to obtain the most satisfactory mixture is within the expected skill and judgment of a mechanic. In re Gibson 5 USPQ 231, 232 (CCPA 1930).

As the Examiner admits, Langer does not disclose the speed of agitator in the stirred tank, however, discloses operating the reaction by a high-speed stirring (example 1). Therefore, it would have been obvious to one having ordinary skill in the art at the time the invention was made to select an appropriate stirring speed such as 300-1000 rpm to well-mix the reaction as taught by Langer to arrive at the instant claimed process.

Claim 19 is rejected under 35 U.S.C. 103(a) as being obvious over Langer, Jr. (4,409,414) in view of Shiraki et al (5,260,500). Langer allegedly discloses a process as discussed above. Langer does not disclose adding thiophene into the catalyst (see the whole patent to Langer for details). However, Shiraki discloses that in a process for producing a linear alpha olefins, it is effective to add to the catalyst a sulfur compound

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such as thiophene to improve the purity of the linear alpha olefins (col. 1, lines 19-28).

It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by adding an amount of thiophene to increase the purity of the product.

Claims 4-12, 20-22, 24-27, and 29 are rejected under 35 U.S.C. 103(a) as being obvious over Langer, Jr. (4,409,414) in view of Young et al (4,855,525).

Specifically, Langer allegedly discloses a process as discussed above but does not disclose using aluminum compounds as called for in claims 4-12, 20, 21, 25, and 26.

However, Young et al. discloses that aluminum compounds such as  $R_3Al_2X_3$ ,  $AlR_2X$ ,  $AlR_3$ , and  $AlRX_2$  are equivalent components for oligomerization catalysts with X being Cl, R being ethyl (the abstract; col. 4, lines 46-51). It would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process by using  $R_3Al_2X_3$  and  $AlR_3$  as the aluminum component for the Langer catalyst since it is expected that using any equivalent aluminum compounds disclosed by Young would yield similar results.

Claims 23 and 28 are rejected under 35 U.S.C. 103(a) as being obvious over Langer, Jr. (4,409,414) in view of Young et al (4,855,525) further in view of Shiraki et al (5,260,500). Specifically, the Examiner contends that Langer and Young disclose a process as discussed above. Neither Langer nor Young disclose adding thiophene into the catalyst (see the whole patent to Langer for details). However, Shiraki is to compensate for this deficiency by allegedly disclosing that in a process for producing linear alpha olefins,

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it is effective to add to the catalyst a sulfur compound such as thiophene to improve the purity of the linear alpha olefins (col. 1, lines 19-28). In the Examiner's opinion, it would have been obvious to one having ordinary skill in the art at the time the invention was made to have modified the Langer process having been modified by Young's aluminum compounds by adding an amount of thiophene to increase the purity the product.

Furthermore, the Examiner contends that Applicant's arguments filed 4/2/04 have been fully considered but they are not persuasive. In the Examiner's opinion, the cited disclosures are directed to mostly very short C<sub>4</sub> to C<sub>8</sub> oligomers and are silent as to producing any major size portion of linear C<sub>10</sub> to C<sub>20</sub> at high yield is not persuasive since this is what is claimed.

The argument that the catalyst used in the present examples is in the form of Zr(OBu)<sub>4</sub>(BuOH) is held not persuasive since the claimed process is not the process in the examples. In other words, applicants do not use a catalyst in the form of Zr(OBu)<sub>4</sub>(BuOH). The argument that the catalysts of the invention have a definite stoichiometry and cannot be generated even by a combination of "in situ" methods described by Langer/Young/Shiraki is not a statement made by applicants without a supporting proof. In other words, applicants do not demonstrate that the catalyst of the prior art cannot be the catalyst of the claimed process.

- **Applicant's Position:**

Applicant respectfully disagrees. On the contrary, the cited references, singly or combined, neither disclose nor even suggest the presently claimed invention. The Applicant asserts that the Examiner has misread the cited references as well as the instant application.

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In the first instance, the rejection of the claims 1-2, 4-7, 9-12, 14, 16, 20-28, and 31 is deemed moot in view of the above cancellation. Claims 3, 8, 15, 17, 18 and 32 have been amended to depend from newly added claim 33.

The new base claim 33 replacing cancelled base claims 1, 20, 26, and 31 is directed to an ecologically friendly process of catalytic oligomerization of ethylene to low molecular weight alpha olefins, comprising the steps of (i) adding to a reactor a catalytic component comprising an alkyl aluminum and a solvent selected from toluene, cyclohexane, and n-octane; (ii) admixing a catalytic component comprising  $Zr(OR)_4$  in an alkanol at a ratio ranging from 1:1.0 to 1:2.3, wherein R is alkyl or aryl; (iii) a further admixing a catalytic component comprising alkyl aluminum halide; and (iv) charging the reactor continuously with ethylene at a pressure ranging from 18 to 35 kg/cm<sup>2</sup> at a temperature ranging from 80 to 140° C. and with a constant agitation.

In addition, amended claim 8, dependent from claim 33, provides that the mole ratio of the aluminum-based catalytic components of process steps (i) and (iii) relative Zr component of step (ii) ranges between 10:1 and 60:1. This ratio is significantly greater than the ratio disclosed in the cited Langer reference wherein the largest mole ratio is limited at 8:1 (Table III).

Applicant respectfully asserts that the claimed process as presently amended is not reasonably predicted by the cited references taken singly or combined. The advantageous process sequence is unobvious over the cited disclosures. No new matter has been introduced with the amendment. The claimed subject matter is fully supported by the instant Specification, especially by the disclosure of the preferred sequence, page 5, lines 4 to 6. It is clear from the description and the examples (especially Example 5) that while the Zirconium(IV) compounds are prepared by reacting with suitable free alkanol, e.g. butanol, the amount of such an alcohol remaining in

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the Zirconium compound when added to the reaction and throughout the reaction is kept within the critical ratios of Zr to alkanol ranging only from 1:1.0 to 1:1.23.

Moreover, Applicant found that the preferred sequence as claimed in newly added base claim 33 advantageously effects the stable multi-component catalyst for the production of LMW alpha olefins from ethylene. The optional thiophene additive as claimed in amended claim 19 is added to reduce chain growth (p.4, l. 7-8), an unobvious property not disclosed or suggested in the cited references.

As presently claimed, Applicant respectfully asserts that the invention provides a surprisingly advantageous process to produce LMW linear oligomers from ethylene using a multi-component catalytic system that controls the size of linear oligomers to a range of four carbon atoms to 24 carbon atoms. This control is unobvious over the cited art which neither discloses nor suggests taken singly or combined. In fact, the cited reference disclosures provide in situ mixings resulting in mostly very short C<sub>4</sub> to C<sub>8</sub> oligomers, and are silent as to producing any major portion of linear C<sub>8</sub> to C<sub>18</sub> at high yield. Moreover, even one of ordinary skill in this art of catalytic conversion of ethylene would know from practical experience that the method of generating the catalytic systems alleged by the combined references would not result in the desirable size of alpha olefins demonstrated by the instant examples. It is also well understood that this art is so unpredictable that the combined cited method would not even remotely arrive at the claimed procedure. The presently claimed procedure on the contrary has been shown to generate a consistently favorable alpha olefin distribution of C<sub>4</sub> to C<sub>18</sub>.

In addition to the previously submitted remarks in traverse of the Examiner's contentions and hindsight suppositions, Applicant asserts that the surprising advantages of the

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instant process is completely different from  $\text{Zr}(\text{OBu})_4$  catalyst used by the primary reference.

It has been unexpectedly found that the proper exchange does not even take place by mere mixing of alcohol and zirconium tetralkoxide as alleged. On the contrary, the catalyst used in the present examples is importantly prepared in the form of, e.g.  $\text{Zr}(\text{OBu})_4(\text{BuOH})$  being in an association at a ratio of 1:0.33 (Example 5). More particularly, the catalyst used in this embodiment of the present invention can be described as  $\text{Zr}(\text{OBu})_4(\text{BuOH})_x$ ; where  $x=0.33$ . This is different from the  $\text{Zr}(\text{OBu})_4$  catalyst used in the Langer reference.

All the above catalyst compositions containing a predetermined free alcohol content within the claimed critical limits, were employed in the oligomerization runs in the invention. Even with the same alkylaluminium but varying the transition metal, different products are formed. It is well understood by the skilled artisan that catalyst systems generated from preformed complexes are quite different in terms of activity and distribution of products in the oligomerization of ethylene (D. Jones et al. 1999 J. Molecular Catalysis 138:37-52).

The very unpredictability of such catalyst systems is illustrated in Run 1 of Table IV of the Langer reference. In short, there simply is no predictability in the catalyst system.

The instant catalyst system also differs from the Langer reference as it does not employ either  $\text{AlEt}_2\text{Cl}$  and/or  $\text{AlEtCl}_2$ . Rather, compounds such as  $\text{Et}_3\text{Al}$  and/or  $\text{Et}_3\text{Al}_2\text{Cl}$  as claimed have been found useful.

Similarly, in regard to the Young reference the compounds taught therein also are known to have significantly different properties than those used in the present invention. For example, the Young reference teaches adducts of  $\text{ZrCl}_4$  with ketones, esters, nitriles, etc. which teach away from the instant method using compounds such as  $\text{Zr}(\text{OR})_4$  (e.g., R = butyl, phenyl). Applicant

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emphasizes that in fact the catalysts of the Young reference and those of the instant invention belong to different class of compounds and give a different reactivity. Consequently, there is no motivation to combine the teachings of the Young reference with those of the Langer reference.

Furthermore, the Shiraki reference differs from the amended claims in that the cited catalyst system is based on the  $ZrCl_4$  system such that the catalyst system, as in Langer, is generated *in situ* where  $ZrCl_4$  and alkylaluminiums are reacted first to provide a bridging complex. Upon adding butanol it bonds  $Zr-Cl$  and  $Al-C_2H_5$  simultaneously. Thus, reactivity of alkylaluminium with alcohol will form undesirable aluminum alkoxides (See, e.g., Kunicki et al, *Studies on the reaction of triethylaluminium with methyl alcohol*, Bull. Polish Acad. Sci. Chem., 33 No. 5 — 6, 1985). Thus, one of ordinary skill in the art would not be able to identify the identity of the active species for the oligomerization reaction.

Finally, completely contrary to the Examiner's reiterated contention which is based on impermissible hindsight, that Shiraki discloses sulfur compounds such as thiophene as unsuitable for allegedly causing contamination of the alpha oligomers. (col 1 lines 29 — 35), the optional use thiophene is based on the advantageous reduction of chain growth. The references are silent on any purity effect of thiophene. The process affords energy saving more moderate temperature ranges and economic solvents much less likely to endanger the environment with toxic waste.

In view of the foregoing amendment and remarks set forth Applicant respectfully asserts that a prima facie obviousness case has not been made and the rejection under 35 USC 103 is deemed improper. Applicant therefore concludes that all claims presently pending in this application are in condition of allowance and are not taught, suggested, or made obvious by the prior art of record.

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### **CONCLUSIONS**

It is respectfully submitted that the subject matter of the pending claims which were rejected under 35 U.S.C. §103(a), are patentable over the references of record. An early notice of allowance is therefore earnestly requested.

Respectfully submitted,

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